

*Sub A1*

CLAIMS

1. A method of automated isotope dilution mass spectrometry comprising
  - providing a sample to be analyzed,
  - spiking at least one enriched stable isotope of an element or specie related to said sample,
  - introducing said spiked enriched stable isotope elements or species into said sample and permitting equilibrium to occur therebetween,
  - subjecting said equilibrated spikes and sample to atmospheric pressure ionization to create ions therefrom,
  - introducing said ions into a mass spectrometer for isotopic ratio determination, and
  - delivering information from said determination to a microprocessor.
2. The method of automated isotope dilution mass spectrometry of claim 1 including
  - employing a liquid sample as said sample.
3. The method of automated isotope dilution mass spectrometry of claim 2 including
  - employing an aqueous solution as said sample.
4. The method of automated isotope dilution mass spectrometry of claim 1 including
  - effecting said spiking on multiple enriched stable isotopic elements.
5. The method of automated isotope dilution mass spectrometry of claim 1 including
  - effecting said spiking on multiple enriched stable isotopic species.
6. The method of automated isotope dilution mass spectrometry of claim 1 including

*Sub A1X*

employing said process to detect levels of one or more contaminants in a sample.

7. The method of automated isotope dilution mass spectrometry of claim 6 including

detecting said contaminants at near instrument detection limits.

8. The method of automated isotope dilution mass spectrometry of claim 6 including

detecting said contaminants at ultra-trace levels.

9. The method of automated isotope dilution mass spectrometry of claim 1 including

after said equilibration but before said ionization, preconcentrating said elements or species.

10. The method of automated isotope dilution mass spectrometry of claim 9 including

effecting said preconcentration through liquid chromatography.

11. The method of automated isotope dilution mass spectrometry of claim 9 including

separating at least one specie of interest by said preconcentration.

12. The method of automated isotope dilution mass spectrometry of claim 1 including

employing said method in qualitative analysis of said elements or species.

13. The method of automated isotope dilution mass spectrometry of claim 1 including

employing said element in quantitative analysis of said elements or species.

*Sub A1X C*

14. The method of automated isotope dilution mass spectrometry of claim 1 including

employing information received by said microprocessor to control operation of portions of said method.

15. The method of automated isotope dilution mass spectrometry of claim 1 including

obtaining said sample from a system being monitored, and delivering a portion of the information received by said microprocessor to said system from which the sample was obtained.

16. The method of automated isotope dilution mass spectrometry of claim 1 including

employing said method to monitor concentration of contaminants in a plurality of wet baths employed in clean rooms in semiconductor manufacture.

17. The method of automated isotope dilution mass spectrometry of claim 16 including

employing said method sequentially on a plurality of said wet baths.

18. The method of automated isotope dilution mass spectrometry of claim 16 including

employing said method simultaneously on a plurality of said wet baths.

19. The method of automated isotope dilution mass spectrometry of claim 1 including

employing a gaseous specimen as said sample.

20. The method of automated isotope dilution mass spectrometry of claim 1 including

employing electrospray ionization as said atmospheric pressure ionization.

- SuJ*  
*A1*  
*Cm*  
*CONFIDENTIAL*
21. The method of automated isotope dilution mass spectrometry of claim 5 including  
effecting said ionization at about 2 to 30 volts.
  22. The method of automated isotope dilution mass spectrometry of claim 4 including  
effecting said ionization at about 200 to 1,000 volts.
  23. The method of automated isotope dilution mass spectrometry of claim 16 including  
employing said method to determine which said bath was the origin of said species or elements.
  24. The method of automated isotope dilution mass spectrometry of claim 1 including  
in effecting said equilibrium equilibrating at least one said spiked enriched stable isotopic specie or element dynamically with a specie or element contained within sample.
  25. Apparatus for automated isotope dilution mass spectrometry comprising

sample receiving apparatus,  
spike introduction apparatus for introducing at least one spiked enriched stable isotope element or specie into said sample for permitting equilibration therebetween,  
an atmospheric pressure ionizer for receiving said equilibrated sample and spiked elements or species and ionizing the same,  
a mass spectrometer for receiving and processing said ions by isotope ratio determination, and  
a microprocessor for receiving information about said determination from said mass spectrometer.
  26. The automated isotope dilution mass spectrometry apparatus of claim 25 including

*Sub*  
*A1*  
*C1*  
*C2*  
*C3*  
*C4*  
*C5*  
*C6*  
*C7*  
*C8*  
*C9*  
*C10*  
*C11*  
*C12*  
*C13*  
*C14*  
*C15*  
*C16*  
*C17*  
*C18*  
*C19*  
*C20*  
*C21*  
*C22*  
*C23*  
*C24*  
*C25*  
*C26*  
*C27*  
*C28*  
*C29*  
*C30*  
*C31*  
*C32*  
*C33*  
*C34*  
*C35*  
*C36*  
*C37*  
*C38*  
*C39*  
*C40*  
*C41*  
*C42*  
*C43*  
*C44*  
*C45*  
*C46*  
*C47*  
*C48*  
*C49*  
*C50*  
*C51*  
*C52*  
*C53*  
*C54*  
*C55*  
*C56*  
*C57*  
*C58*  
*C59*  
*C60*  
*C61*  
*C62*  
*C63*  
*C64*  
*C65*  
*C66*  
*C67*  
*C68*  
*C69*  
*C70*  
*C71*  
*C72*  
*C73*  
*C74*  
*C75*  
*C76*  
*C77*  
*C78*  
*C79*  
*C80*  
*C81*  
*C82*  
*C83*  
*C84*  
*C85*  
*C86*  
*C87*  
*C88*  
*C89*  
*C90*  
*C91*  
*C92*  
*C93*  
*C94*  
*C95*  
*C96*  
*C97*  
*C98*  
*C99*  
*C100*

a sample analyzer for analyzing said sample and delivering sample analysis information to said microprocessor.

27. The automated isotope dilution mass spectrometry apparatus of claim 26 including

a controller for receiving information processed by said microprocessor and providing feedback to other portions of said apparatus.

28. The automated isotope dilution mass spectrometry apparatus of claim 25 including

sample modification apparatus for altering characteristics of said sample responsive to signals from said controller prior to the sample entering said atmospheric pressure ionizer.

29. The automated isotope dilution mass spectrometry apparatus of claim 28 including

said controller coordinating operation of said sample receiving apparatus, said spike introduction apparatus, said chemical modification apparatus, said atmospheric ion generator and said mass spectrometer.

30. The automated isotope dilution mass spectrometry apparatus of claim 29 including

solution handling apparatus interposed between said chemical modification apparatus and said atmospheric ion generator, and

at least one chromatograph operatively associated with said solution handling unit for preconcentrating said equilibrated sample and spiked enriched stable isotope elements or species prior to delivery to said atmospheric ion generator.

31. The automated isotope dilution mass spectrometry apparatus of claim 30 including

said chromatograph including at least one chromatograph selected from the group consisting of a liquid chromatograph and a gas chromatograph.

*Sub A1X*

32. The automated isotope dilution mass spectrometry apparatus of claim 30 including

said sample receiving apparatus having a first outlet conduit in communication with said spike introduction apparatus which in turn has an outlet conduit in communication with said chemical modification apparatus and a second conduit in communication with said chemical modification apparatus whereby a sample may be admixed with said spiked enriched stable isotope elements or species in effecting quantitative analysis and may be introduced directly into said chemical modification apparatus without admixture with said spiked elements or species for effecting qualitative analysis.

33. The automated isotope dilution mass spectrometry apparatus of claim 25 including

said atmospheric ion generator being an electrospray ionizer.

34. The automated isotope dilution mass spectrometry apparatus of claim 25 including

said atmospheric ion generator being structured to operate at a first voltage when effecting ionization of multiple enriched stable isotopic elements and a lower second voltage when ionizing multiple enriched stable isotope species.

35. The automated isotope dilution mass spectrometry apparatus of claim 34 including

said first voltage being about 200 to 1,000 volts and said second voltage being about 2 to 30 volts.

36. The automated isotope dilution mass spectrometry apparatus of claim 25 including

a system interface for receiving information from said microprocessor and providing feedback to the system being monitored.

37. The automated isotope dilution mass spectrometry apparatus of claim 36 including

said system interface having a warning capability if the concentration of a monitored contaminant approaches a tolerable upper limit thereof

and an alarm capability if the concentration of said contaminant reaches or exceeds the tolerable upper limit.

38. The automated isotope dilution mass spectrometry apparatus of claim 25 including

said atmospheric ion generator being atmospheric pressure chemical ionizer.

*Sub  
Al  
and*